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Abstract

This paper presents results regarding the potential of lignosulfonate (LS) to control the swelling of expansive soil. One-dimensional swell tests were performed on untreated and LS-treated remoulded samples of Australian expansive soil from the state of Queensland. The test results indicated that LS has significant influence on the swelling behaviour of this expansive soil. The results were compared with those of identical cement-treated soil samples, and it was found that LS could be a economical and environmentally friendly alternative to traditional alkaline additives. In addition, the behaviour of LS-treated specimens during repeated freezing and thawing cycles was measured. The results indicated significant improvement in the percentage mass loss in LS-treated specimens compared with cementtreated specimens. The microstructural analysis of the untreated and the LS-treated samples showed soil surface area reduction in the treated specimens, which in turn reduced the affinity of the specimens towards moisture uptake, resulting in a reduction in the swell potential of the otherwise expansive soil. The use of LS as a novel non-traditional stabiliser for expansive soil appears to be a viable solution in view of the sustainable use of waste by-products and green construction.

Keywords

lignosulfonate, treated, swelling, expansive, behaviour, soil

Disciplines

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The Swelling Behaviour of Lignosulfonate Treated Expansive Soil

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Abstract

This paper presents results on the potential of Lignosulfonate (LS) in controlling the swelling of expansive soils. One dimensional swell tests were performed on untreated and LS treated remoulded samples of an Australian expansive soil from the state of Queensland. Test results indicated that LS has significant influence in the swelling behaviour of this expansive soil. The results were compared with those of cement-treated identical soil samples, and it was found that LS could be a resourceful and environmental friendly alternative to traditional alkaline additives. In addition, the behaviour of LS treated specimens during repeated freezing and thawing cycles were measured. The results indicated significant improvement in percentage mass loss in LS treated specimens compared to cement treated specimens. The microstructural analysis of untreated and LS treated samples showed soil surface area reduction in treated specimens, which in-turn reduced the affinity of the specimens towards moisture uptake, thus the reduction in swell potential of the otherwise expansive soil. The use of LS as a novel non-traditional stabilizer for expansive soil appears to be a viable solution in view of sustainable use of waste by-products and green construction.

List of notations

SEM	scanning electron microscope
EDS	energy dispersive spectroscopy
FTIR	fourier transform infrared
¹ H NMR	hydrogen nuclear magnetic resonance
BET	Brunauer–Emmett–Teller analysis
XRD	x-ray diffractometer
OMC	optimum moisture content
MDUW	maximum dry unit weight
LS	lignosulfonate

Keywords: Expansive soil, chemical stabilization, lignosulfonate, microstructure

Introduction

The threat posed to public safety and the potential for destruction of properties by expansive soils is growing rapidly. Worldwide, billions of dollars are budgeted in controlling the adverse effects of shrink – swell phenomenon of expansive soils (Jones and Jefferson, 2012). For example, in Australia, expansive soils cause several millions of dollars in damage to infrastructure annually (Richards, 1990).

In the past, researchers (Puppala et al., 1999; Cameron, 2006) have developed various techniques to minimise the adverse effects of expansive soils on infrastructures. Chemical stabilisation using traditional alkaline admixtures (e.g. cement and lime) is the most common technique adopted in controlling the shrink-swell behaviour of expansive soils. Traditional admixtures reduce shrink-swell potential and increase the workability of expansive soils. However, the use of these alkaline stabilizers may pose intrinsic environmental and occupational health/safety problems. For example, the pH of soil increases upon treatment, and this high pH could cause corrosion problems in buried utilities (Rao and Thyagaraj 2003). In addition, the use of these alkaline stabilizers presents adverse implications in terms of cost and emission of cancerous particulate matter, greenhouse gasses and consumes about 2% of global energy in their production processes (e.g. Boynton, 1980; Albino *et al.*, 2011).

At present, the use of industrial by-products (non-traditional) as alternative stabilizing agents has become increasingly popular (e.g. Puppala et al. 1999; Tingle and Santoni, 2003; Indraratna et al. 2010; Vinod et al. 2010; Athukorala et al 2013). Many of these non-traditional stabilizers have shown to require much less quantity for soil stabilization compared to traditional admixtures or superior durability properties and/or reduced curing time (Indraratna *et al.*, 2010; Harichane, 2011). Despite these research achievements, unfortunately, there exist no standard protocols to aid practitioners to identify non-traditional stabilizers that deliver enhanced performance and those that do not. From the above context, this study focuses on understanding

the potential of a non-traditional admixture called LS in controlling the swelling of expansive soil.

LS is a by-product of the sulphite pulping process in paper manufacturing. It is a non-corrosive, non-toxic, and environmental friendly chemical. As the second most abundant natural polymer in the world (Thakur et al., 2014; Gandini and Belgacem, 2008), lignin based compounds have often tempted researchers to study their potential for use in novel applications. For instance, Athukorala et al., (2013) described a theoretical model to capture the rate of erosion of a silty sand based on the principle of conservation of energy. The proposed equation is based on the shear stress-strain characteristics, mean particle diameter, mean flow velocity, and the packing arrangement of particles. The model results were reported to be in agreement with experimental data. In the petroleum industry, LS has been used in the production of oil drilling muds. It is reported that the presence of LS reduced the viscosity of the mud thus, reducing the energy requirements needed for oil drilling (Orszulik, 2013). Indraratna *et al.* (2010) investigated the internal erosion behaviour of LS amended silty soil by varying the amount of LS and cement from 0.2 to 0.6% dry weight of soil. The LS amended soil reduced the coefficient of soil erosion and significantly increased the critical shear stress in comparison to soil treated with cement. Tingle and Santoni (2003) compared different traditional (e.g. cement and hydrated lime) and non-traditional admixtures including LS to evaluate the strength of low-and high- plasticity clay soils. The above study reported that LS provided excellent waterproofing for the expansive soils compared to the rest of the stabilizers. To the authors' knowledge, currently there is no information on the potential of LS in controlling the swell behaviour of expansive soils. This study intends to contribute to this research gap. Moreover, freezing and thawing tests were carried on LS treated soil to understand the durability of the treated soil for the potential application in temperate regions of the world.

The results of the study are complimented with microstructural analysis of untreated and LS treated samples using x-ray diffraction (XRD), scanning electron microscope (SEM) and Brunauer Emmett and Teller (BET) surface area analysis tests. LS was also examined using energy dispersive spectroscopy (EDS), fourier transform infrared (FTIR) spectroscopy, ultraviolet spectroscopy (UVS) and nuclear magnetic resonance (NMR) spectroscopy thus, a chemical structure for the otherwise proprietary chemical stabilizer (LS) is proposed.

Materials and specimen preparation/test methods

The soil used in this study is a highly plastic expansive clayey soil which was obtained at 25 – 70cm depth at Gunnedah (Queensland, Australia). The retrieved soil samples were placed in sealed plastic bags, and transported to the laboratory for preparation and testing. Extensive laboratory tests were carried out to classify the soil and the results (Table 1). This soil could be classified as expansive on the basis of its plasticity, percent swell, shrinkage limit and activity. The high liquid limit (>50%) and high plasticity index (51%) support the typical characteristics of a montmorillonitic expansive soil (AS 1726-1993). The percent swell of approximately 6%, classifies this soil as “high” expansivity following the classification proposed by Seed et al. (1962) (i.e. percent swell = 5 - 25% under 7kPa surcharge). The soil’s shrinkage limit is 9% which agrees with the same class of expansiveness (7 – 12%) (Holtz and Gibbs, 1956). Furthermore, the soil activity of 1.42 also supports the soil’s expansiveness (Skempton, 1953).

The averaged elemental composition of the expansive soil as obtained from EDS technique is also shown in Table 1. The presence of Si, Al, Mg, Ca, Fe, and Na indicated the existence of montmorillonite minerals in the soil. XRD diffractograms analysis with *Siroquant* software showed that the soil contained a mixture of 36.0% clay of which montmorillonite = 10.4%, montmorillonite-illite mixed mineral = 0.6%, montmorillonite-chlorite mixed mineral = 8.5%; illite content = 4.4%; kaolinite fraction is 12.7% and quartz = 25.3% by percentage mass. This is consistent with the classification of “high” expansiveness based on the physical index

properties. This is consistent with the classification of “high” expansiveness based on the physical index properties.

Specimen preparation

The clayey soil collected from the field was oven-dried at 105⁰C to constant mass, pulverized and sieved. The materials passing the 1.18mm sieve were collected as test soil. An aliquot of the dry soil was thoroughly hand-mixed with desired moisture content. The mix was then placed in sealed double plastic bags and allowed to mellow for 24hr to permit moisture and LS (for treated samples) equilibration prior to specimen preparation. Swell test specimens were statically compacted directly into consolidometer rings of diameter 50mm x 20mm height using a specially designed mould at a rate of 1mm/min to desired density. Freeze-thaw specimens were also compacted in similar manner into 115mm height x 105mm diameter mould to predetermined density. It should be noted that LS treated specimens were prepared in the same manner but with the addition of 2% LS by dry weight of soil. All compacted specimens were cured for 7 days prior to each test to allow for possible interaction between soil minerals, water, and, LS stabilizer. To compare the performance of LS in relation to traditional admixtures, additional but identical specimens were prepared with 2% cement admixture. Cement was selected because it induces lesser environmental impact compared to lime admixture and past research studies have shown that at reasonably low application rates (1 - 2% by weight) both lime and cement admixtures give comparable results (Chen 1988).

Swell and durability test methods

The percent swell test for untreated and LS treated specimens were carried out in accordance with ASTM D4546 but with a 7kPa seating pressure. The specimens were placed in oedometer equipment and inundated with deionised water and allowed to swell for typically 10-21 days. At the end of one-dimensional swell tests, volumetric-shrinkage tests were conducted in accordance

with the procedure proposed by Briaud (1998). The dimensions and weights of the swollen specimens were measured and monitored in 24hour intervals as the specimens were allowed to dry at room temperature to constant mass. The specimens were then transferred into an oven at 105 °C for 3 days. The volumetric shrinkage was calculated from the above data.

The “wetting-after-loading tests on multiple specimens” method (ASTM D4546) was employed in determining the swell pressure of untreated and LS treated samples. The method involved applying increasing vertical load on inundated specimens. At each seating load, the corresponding swell or hydro-compression was measured and recorded. The data obtained was used to plot “swell/hydro-compression versus applied pressure” relation. The swell pressure was reported as the averaged maximum applied stress required to preventing the specimens from swelling or compressing i.e. the point at which the plot-line intercept the zero mark on the vertical axis.

Freeze-thaw durability tests were performed to simulate the effect of seasonal variation on mass loss, adsorbed moisture variation and volumetric changes in untreated and LS treated specimens in accordance with ASTM D 560. In addition, the effect of LS stabilizer on soil pH was evaluated with a pH meter (AS 1289.4.3.1) and compared to cement stabilized soil sample.

Lignosulfonate (LS); proposed chemical structure

FTIR spectroscopy at mid-infrared region (600–4000 cm^{-1}) was applied on LS stabilizer and the absorption bands were matched with standards given in Coates (2000) (Fig 1a). The characteristic bands at 3200-3500 cm^{-1} , 2920 cm^{-1} , 2842 cm^{-1} , 1455-1591 cm^{-1} , 1038 cm^{-1} , and 1115-1173 cm^{-1} , indicated the presence of hydroxyl group (-OH), methylene group (CH_2), methoxyl group (OCH_3), aromatic benzene ring ($\text{C}=\text{C}$), alcohol ($\text{C}-\text{O}$); [note that alcohols have IR absorptions linked with both C-O stretching and the O-H vibrations (Coates, 2000)] and the sulfonate group (SO_3^-), respectively. The UVS confirm the presence of phenyl groups in the

stabilizer. The ^1H NMR (hydrogen nuclear magnetic resonance) spectrum of LS confirmed the FTIR results and identified the position (ortho-, meta- or para-) of functional groups. The spectrum also reveal the presence of sulphur group species attached to an organic chemical and the presence of hydroxyl group at 6-12ppm due to intermolecular hydrogen bonding (Zhao *et al.*, 2005). The major elemental constituents of LS were also determined using EDS and are presented in Table 2. The calcium composition quantitatively supersede other elements present, which implies that calcium is the dominant salt present in the LS, thus the LS is a CaLS. Based on the data obtained from the above analytical techniques, a chemical structure of the LS stabilizer is proposed (Fig 1b).

Results and discussion

Effect of LS on engineering properties of expansive soil

Fig 2a shows the variation of liquid limit (LL) and plasticity (PI) characteristics based on the percentage of LS admixture. For comparison, the effect of cement content on the consistency limits of the same soil is presented. The addition of LS resulted in a decrease in the LL (91 to 76%), and a slight increase in the plastic limit (40 to 44%), prompting a significant 37% reduction in soil plasticity index (51 to 32%) at 2% addition. The change in PI could be attributed to the transformation of the soil mineral lattices that contribute to plasticity, because the average size of the soil particles became larger, transforming soil into a more silty state. This observation was supported by particle size distribution analysis of untreated and treated soil (Fig 2b). The content of clay size particles decreased from 36% to 20% probably due to aggregation, which in turn decreased the plasticity of the soil. However, increasing LS content beyond the 2% threshold resulted in adverse soil characteristics. For example, at 4% application rate, the LL decreased to 80% only, with a corresponding decrease in plastic limit to 41%. Hence, 2% LS content was considered as the optimum in this study.

For cement treated soil, both the LL and PI decreased with increasing cement content. The LL (91%) of the untreated soil decreased to 60% while the plastic limit increased from 40% to 46%, at 7% cement addition which resulted in PI reduction from 51% to 14%. Sarkar et al. (2012) attributed such LL behaviour to the greater quantity of water required for the cement treated soil to make it fluid while the increase in plastic limit for cement treated soil was attributed to the greater amount of water required to change the soil's plastic state to semisolid state.

Though, the PI of cement treated soil is significantly lower than the LS treated sample, LS prompted a significant 41% plasticity reduction in the soil. Thus, LS could also effectively improve the plasticity property of a “low” expansive soil. It was also observed that the pH of LS treated soil remain practically unchanged (from 7.43 to 7.17 after treatment). This is indeed an outstanding advantage of LS stabilizer over its traditional counterparts because treating the same soil with 2% cement increased the alkalinity from 7.43 to 9.19).

Shrink/swell behaviour

The effect of LS content on percent swell with time is shown in Fig 3, whereas, Fig 4b shows the effect of initial moisture content on the shrink/swell behaviour of specimens before and after LS treatment. The variability in test results is evident despite the identical manner in which specimens were prepared. When treated with 2% LS, the percent swell reduced from 6% to about 4.6%, which is a significant improvement in swell behaviour of the soil. As earlier stated, the improvement in the PI of the treated soil implies that the wettability of the soil decreased thus, the decrease in the percent swell. Saride et al. (2013) showed that PI is a good indicator of shrink/swell behaviour of soil; the lower the PI the lower the shrink/swell behaviour and vice versa.

Similarly, the percent swell (Fig 3) decreased with increasing content of LS until a threshold value of 2% by dry weight of soil is reached. Beyond this dosage, the percent swell increased

with increasing admixture content. This anomalous behaviour has been attributed to mutual repulsive forces between the charged sections of the admixture (Sarkanen and Ludwig, 1971).

After swelling was complete, a shrinkage test was conducted and data obtained was used to determine the volumetric shrinkage of specimen at various moisture contents (Fig 4). The shrinkage remained fairly the same within a moisture content range of 15 to 60%. At OMC, the volumetric shrinkage of untreated and 2% LS treated specimens is 11.0% and 13.5%, respectively. The effect of 2% LS content on volumetric shrinkage is not as pronounced as the effect on swell behaviour. However, the shrinkage limit of treated sample increased slightly from 9% to 11% after treatment. The slight improvement in shrinkage limit value could be as a result of the stabilization mechanism of LS in that, the entrapped moisture in treated soil may be more tightly bonded in the interlayers of clay minerals.

The magnitude of the swell pressure of the soil was measured to be 105kPa but decreased to 84kPa after 2% LS addition (Fig 5). For cement-treated specimens, the reduction in swell pressure is similar to that of 2% LS treatment (82kPa). Within applied surcharge pressure of about 80 to 280kPa, the treated specimens compressed more than the untreated sample. This could be associated with the high intrinsic property of the untreated sample. In other words, the untreated sample has greater tendency to swell than the treated samples under the same surcharge pressure. However, at surcharge pressure of about 300kPa, the compression is the same for both untreated and treated samples. The surcharge pressure at which the compression of untreated sample equals that of the treated samples is referred herein as the critical pressure. The significance of the critical pressure is that it equals the pressure beyond which the compressive behaviour of untreated and treated specimens does not change with time.

Curing time dependent swell behaviour of LS treated expansive soil

The effect of curing time on percent swell of the soil was evaluated by conducting swell tests on soil – water - 2% LS mix, compacted at OMC and cured for periods of 7, 14, 54, and 180 days. For comparison, identical cement treated specimens were prepared and tested accordingly. The rate of swell depends on curing time in 2% LS treated soil, though the magnitude at the end of the swell regime is independent of such time (Fig 6). The rate of swell decreases with increasing curing period. After 10 minutes of swell test, it was observed that the untreated soil would have almost completed its primary swell (at 4%) whereas, for the 180 days cured sample, the percent swell was significantly low (at 0.4%). The swell rate dependency on curing time could mean that the stabilization processes of adsorption, coating and subsequent aggregation of particles by LS stabilizer is time-dependant under laboratory conditions.

On the other hand, the cement treated specimen exhibited insignificant variation in swell rates regardless of curing time, though the reduction in percent swell is more significant, accounting for 33% i.e. from 5.9% to 3.9% (Fig 7). The independent behaviour of the rate of swell with curing time implies that cement stabilization processes on expansive minerals are completed within 7 days after treatment. Although, at higher application rates (e.g. 4 or 8%), soil pH could increase to 12.4 which could initiate pozzolanic reactions in which case the above statement might not be valid.

Freeze-thaw durability test

The durability conditions of untreated and 2% LS treated specimens under repeated freezing and thawing cycles were measured by percentage mass loss, percentage moisture variation and percentage volumetric change. For the purpose of comparison, 2% cement stabilized identical specimens were prepared and tested. The samples were prepared at optimum moisture content (standard Proctor) and compacted into a 104 mm diameter by 100mm height mould and then cured for 7 days prior to freeze-thaw cycles. The freezing-and-thawing cycle for the tests consisted of freezing the specimens at -4°C for 24 hours and thawing for 24 hours at room

temperature of 20°C with 99% humidity. Freezing a specimen for 24 hours and thawing it for another 24 hours represents one cycle. Samples were predetermined to undergo 12 cycles of freezing and thawing in 24 days.

During the freezing stage, as the temperature drops below 0°C, the soil moisture starts to freeze and results in the formation of ice crystals which subject the specimen to change its volume (Konrad, 1989) (Fig 8). Throughout the freeze–thaw tests for untreated and 2% LS treated specimens, both samples developed cracks around the entire surfaces causing the specimens to break off at the lower and upper ends. As the test continued, these cracks became more significant and widespread, resulting in significant spalling and eventual break down of the specimens. Cement treated specimen experienced the highest level of mass loss, as a result, the durability test was stopped after 6 cycles (Fig 8). The degraded post-test images of specimens are also shown in Fig 8. It is interesting to note that the number of cycles at which deterioration of the specimens began was considerably affected by the presence of LS.

Through visual inspection, less spread of surface spalling was observed in LS treated specimens from the 1st to the 12th freeze thaw cycle, demonstrating a substantial improvement in resistance to degradation. At the end of the 12th cycle, the untreated soil lost approximately 7% of its mass while the LS treated specimen experienced mass loss of 3.4% only. In contrast, 2% cement stabilized soil specimen experienced rapid deterioration, losing as much as 17.8% at the end of the 6th cycle (end of test). This substantial mass loss could compromise the strength of cement stabilized soil (Chamberlain et al., 1990). Chamberlain et al., (1990) reported that 10–15% mass loss at the end of 12 cycles of freezing and thawing insignificantly affects the strength of soil close to surface. The 2% LS treated sample is well below this range of mass loss and it is expected that the strength of LS treated soils may not be affected due to mass loss with changing temperature.

In terms of volume change, the untreated specimen showed significant variation in volume from the 1st cycle to the 6th cycle, while cement treated specimen exhibited the least change. The maximum volume change for all specimens occurred at the end of the 2nd cycle. The volume increased by almost 15.9% in untreated sample as opposed to 11.2% and 6% for 2% LS and 2% cement treated specimens, respectively (Fig 9). The volume change in each subsequent cycle was observed to be less than the preceding cycle and this trend continued until about the 9th cycle for untreated and LS treated specimens. Beyond this cycle, repeated freezing and thawing conditions showed no significant effect on specimen volume, indicating that the specimens have reached their equilibrium state after the 9th cycle. At this state, the maximum volume change for untreated soil is 10.6% as opposed to 9.2% in LS treated specimen. Chen (1988) referred to the progressive decrease in volume change towards the equilibrium state as the “fatigue” of volume change as a result of macrostructural softening due to microstructural expansion.

For moisture variation determination in specimens, the moisture content at the end of each cycle was measured. A unique relation existed between moisture content variation and volume change in specimens. It was observed that the moisture content was proportional to volume change (Fig 10). Particularly significant is the reduction in the adsorbed moisture content in 2% LS treated soil. At the end of the 9th cycle, the gravimetric moisture content of each sample was determined. Consequently, the adsorbed moisture content was least in the cement treated specimen (41%) followed by LS treated specimen (47%) and then the natural specimen (50.9%). It is most likely that the adsorbed hydrophobic organic molecules of LS contributed to restricting water penetration into the soil body.

Microstructural analysis of LS treated Soil

XRD Analysis

XRD was employed in this study to assess the changes in soil mineralogy due to LS addition. The XRD diffractograms of the untreated and 2% LS treated oriented specimens are presented in Fig 11. The diffractograms were compared for the existence of new peaks (new minerals) in treated samples. The distance of the separation of atomic planes (d-spacing) was also investigated to understand the mineralogical alterations in specimens. The peaks located at $2\theta = 21^\circ$, 27° , 37° and 50° are signature peaks of quartz whereas, the peaks at approximately $2\theta = 5.6^\circ$, 18° , 28.5° , 35.5° , 42° and 54° matched with known montmorillonite spectra contained in the database. For LS treated specimen, the montmorillonite peaks are invariably broader, reflecting the much smaller size of crystallite as a result of LS addition. A comparison of untreated and 2% LS treated diffractograms indicated a shift at the 001 montmorillonite peak to the left (an increase in d-spacing) for treated sample, suggesting that LS intercalated the inner layers of the montmorillonite lattices and forced it to expand initially. Thus, the peak at $2\theta = 5.6$ shifted to 5.39 which corresponded to an increase in the d-spacing from 15.77\AA to 16.38\AA , respectively. In addition, the montmorillonite patterns at $2\theta = 5.6^\circ$, 18° , 28.5° , 35.5° , 42° and 54° either broadened or completely disappeared in the treated sample. The changes in the peaks at these scan angles suggested that LS altered or destroyed the montmorillonite lattices. The peak intensities of other minerals such as quartz, kaolinite and illite at $2\theta = 50.5^\circ$, 25° and 55° also decreased, respectively, after treatment. However, XRD patterns did not show intercalation of LS into the inner layers of these minerals, signifying that only peripheral adsorption of LS occurred on the surfaces of these minerals. The likely reason is the large molecular weight of LS polymer as compared to the very small inner layer spacing in non-expansive soil minerals. The adsorption processes subsequently coated the soil mineral lattices to form flocs and restricted the characteristic diffraction of the atomic planes especially in montmorillonite leading to restricted movement of water into the entire clay matrix hence, the reduced swell behaviour. No significant new peak was observed after treatment with LS, a subdominant peak at about $9^\circ 2\theta$ was seen which corresponds to the presence of additional illite minerals (Fig 11). This implies

that the illite mineral content increased after LS addition which contributed to the reduction in the intrinsic swell properties of this otherwise expansive soil.

SEM analysis

This analytical technique was employed to qualitatively identify the microstructural development in the matrix of the stabilized soil specimens at x3000 magnification (Fig 12). The fabric of the untreated sample has a dense clay matrix with no appearance of aggregations and connectors, thus the voids are more visible. The micro-pores are unevenly distributed and their dimensions show an erratic arrangement.

On the other hand, the 2% LS treated soil micrograph exhibited marked changes in morphology. The untreated soil was transformed from a particle based structure to a more integrated composite with larger but fewer inter-aggregates dominate pore spaces. The formation of larger inter-aggregate pore spaces resulted in a decrease in the number and volume of intra-aggregate pore spaces hence, restricted the movement of water molecules into the entire clay matrix (reduced percent swell) (Monroy et al., 2005). This could be a reason why the measured moisture content of 2% LS treated samples at the end of swell test is 47% as opposed to 51% for untreated soil. The theory that aggregation of soil particles forms part of the stabilization mechanism in LS treated soil was further probed with the BET surface area analysis technique as discussed below.

BET analysis

BET surface area analysis technique was used to assess changes in the soil's external surface area attributable to 2% LS admixture. BET nitrogen adsorption surface area results indicate $58.88\text{m}^2/\text{g}$ and $32.93\text{m}^2/\text{g}$ for untreated and LS treated samples, respectively. The addition of LS stabilizer led to reduction in the surface area due to aggregation of particles. The implication is that the soil will adsorbed less moisture due to the reduced number and volume of intra-

aggregate pore spaces (Fig 12) which in turn contributed to the decrease in the percent swell of the soil.

Possible stabilization mechanism based on the microstructural analysis

On the basis of XRD analysis, LS underwent basal and peripheral adsorption on the surfaces of the montmorillonite minerals and only peripheral adsorption on non-expansive soil minerals. This resulted in smearing and subsequent aggregation of soil particles as seen from SEM micrographs and BET surface area analysis data. The reduction in the number and volume of intra-aggregate pore spaces compensated by the formation of larger but fewer inter-aggregate pores as a result of LS adsorption restricted the ingress of water into the entire clay matrix hence; the decreased percent swell of the treated soil.

In addition, to the above mechanisms, the disappearance or reduction in signature peak heights of soil minerals (Fig 11) and Peak broadening at 001 montmorillonite suggested reduction in crystallite size of the soil minerals ($2\theta = 20^\circ$, 27° and 48°) due to the smearing mechanism of the admixture. The encapsulation of soil particles resulted in the reduction in peak heights of minerals. In summary, LS stabilized the expansive soil by changing the characteristics of the soil crystals via basal and peripheral adsorption as opposed to significant chemical reactions because of the absence of any remarkable change in mineralogy of 2% LS treated soil. The implication is that the soil plasticity decreased which in turn reduced the shrink-swell potential of the otherwise expansive soil. However, depending on the mineralogical composition of a soil, the authors believe that cation exchange could be a possible additional reaction mechanism of the admixture as divalent and trivalent ions (e.g. Al^{3+} , Mg^{2+} and Ca^{2+}) in LS could readily replace exchangeable monovalent ions (e.g. Na^+ , K^+ and Li^+) in the expansive soil mineral surfaces. Advanced cation exchange testing (Ammonium Acetate Method) is recommended for the verification of the above statement.

Conclusions

To evaluate the potential effectiveness of LS in stabilizing a remoulded expansive soil, swelling and durability (i.e. freeze-thaw) tests were conducted and complemented with microstructural analysis. On the basis of the obtained experimental results, the following conclusions are drawn:

- The results suggest that the optimum LS content to provide a better improvement on the percent swell is 2% by dry weight of the soil and the improvement continues with increasing curing time for up to 180 days. In terms of specimens' resistance to freeze-thaw durability conditions, the LS stabilized specimen exhibited the most significant improvement with respect to percentage mass loss.
- The PI, percent swell, swell pressure, and volumetric shrinkage improved from 51% to 32%, 5.9% to 4.6%, 105kPa to 84kPa, and 9% to 11%, respectively. The rate of swell of 2% LS treated soil also decreased with increasing curing time. These changes are caused by the binding together of soil minerals by LS admixture through basal and peripheral adsorption, which consequently restricted moisture infiltration into the entire soil body.
- Of interest is the fact that soil pH does not change (7.43 to 7.13) upon LS addition which is in direct contrast with cement admixture that considerably increased soil pH from 7.43 to 9.19 at 2% application. Such pH level could be detrimental to buried steel structure or flora and fauna.
- XRD diffractograms of 2% LS treated expansive soil revealed basal and peripheral adsorption of LS by expansive minerals but only peripheral LS adsorption by non-expanding soil minerals resulting in coating and aggregation of soil particles as observed during BET and SEM analyses. The coating effect caused a reduction in crystallite size of the expansive mineral (broadening of 001 montmorillonite peak). The proposed stabilizing mechanisms of LS as identified in this study involved initial

interlayer expansion during LS intercalation, subsequent moisture entrapment and encapsulation of clay minerals; i.e. changes in the crystallographic properties of the soil minerals. In other words, the stabilizing mechanisms are of physical means, involving changes in the crystallographic properties of the soil rather than significant chemical interactions.

The significant reduction in swell behaviour achieved in the laboratory may not be achieved in field applications where the conditions are much more complex, but the fact remains that LS reduced the adsorbed moisture content in treated soil and hence, reduced the swell potential of the soil. This has a lot of positive implications to the management of expansive soil related infrastructural damage. The authors therefore, recommend the use of this by-product as a stabilizer for controlling swell potential and at the same time solve a waste disposal problem for the paper manufacturing industry. It is suggested that field trials should involve the application of LS in the traditional format currently used for lime and cement admixtures.

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References

- Albino V, Dangelico RM, Natalicchio A, and Yazen DM (2011) Alternative Energy Sources in Cement Manufacturing: A systematic review of the body of knowledge. *Network for Business Sustainability, University of Western Ontario, Ontario, Canada.*
- Australian Standards 1289 (1997) Methods of testing soils for engineering purposes. Method 4.3.1: Soil chemical tests-Determination of the pH value of a soil-Electrometric method.
- Australian Standards 1726 (1993) geotechnical site investigation, Classification and identification, third edition.
- ASTM (1996) D560 Test Methods for Freezing-and-Thawing Tests of Compacted Soil- Cement Mixtures, West Conshohocken, PA, USA.
- ASTM (2008) D4546 Standard test method for One-Dimensional Swell or Collapse of Cohesive Soils, West Conshohocken, PA, USA.
- Athukorala R, Indraratna B and Vinod JS (2013) Modeling the internal erosion behavior of lignosulfonate treated soil. In C. L. Meehan, D. Pradel, M. A. Pando & J. F. Labuz (Eds.), *Geo-Congress 2013 (1872-1881)*. United States: American Society of Civil Engineers
- Boynton RS (1980) *Chemistry and Technology of Lime and Limestone*. 2nd Ed., Wiley, New York.
- Briaud JL (1998) Shrink Test for Predicting Heave and Shrink Movements. *Proceedings, ASCE Texas Section Fall Meeting*, Dallas, Texas.
- Cameron DA (2006) Role of vegetation in stabilizing highly plastic clay subgrades, *International Conference on Railway Track Foundations*, eds. Ghataora, G. S. and Burrow, M. P. N., Birmingham, UK, September 11-13, Birmingham University Press, 165-186.
- Chamberlain EJ, Iskander I and Hunsiker SE (1990) Effect of freeze-thaw on the permeability and macrostructure of soils. *Proceedings International Symposium on Frozen Soil Impacts on Agriculture, Range, and Forest Lands*. Special Report 90 – 1: 145–155.
- Chen FH (1988) *Foundations on expansive soils*. Elsevier Science Publishers, Amsterdam, The Netherlands.
- Coates J (2000) Interpretation of Infrared Spectra: A Practical Approach in *Encyclopedia of Analytical Chemistry* R.A. Meyers (Ed.) 10815–1083, Wiley, Chichester.
- Gandini, A., Belgacem, M. (2008). Lignin as Components of Macromolecular Materials. In *Monomers, Polymers, and Composites from Renewable Resources*; Eds.; Elsevier: Oxford, UK, 243–273.
- Harichane K, Ghrici M and Kenai S (2011) Effect of curing time on shear strength of cohesive soils stabilized with combination of lime and natural pozzolana. *International Journal of Civil Engineering*, 9(2): 90-96.
- Holtz WG and Gibbs HJ (1956) Engineering properties of expansive clays. *Transact. ASCE* 121: 641-677.

- Indraratna B, Mahamud M, Vinod JS and Wijeyakulasuriya V (2010) Stabilization of an erodible soil using a chemical admixture, In Bouassida, M, Hamdi, E & Said, I (eds), *ICGE'10: Proceedings of the 2nd International Conference on Geotechnical Engineering*: 45-54.
- Jones LD and Jefferson I (2012) Expansive soils, *Institution of Civil Engineers Manuals Series*, ICE Publishing, 1-46.
- Konrad JM (1989) Effect of freeze-thaw cycle on the freezing characteristics of clayey silt at various overconsolidation ratios. *Canadian Geotechnical Journal*, 26(2): 217-226.
- Monroy R, Zdravkovic L and Ridley A (2005) Fabric Changes in Fabric Changes in Compacted London Clay Due to Variations in Applied Stress and Suction. *Experimental Unsaturated Soil Mechanics Springer Proceedings in Physics*, 112: 41-48.
- Orszulik, S.T. (2013). Environmental Technology in the Oil Industry, Springer Science and Business Media, Netherlands, Also available at <https://books.google.com.au/books?id=QDjpCAAAQBAJ>, accessed on August 10 2015.
- Puppala AJ, and Hanchanloet S (1999) Evaluation of a new Chemical (SA-44/LS-40) Treatment Method on Strength and Resilient Properties of a Cohesive Soil. *Technical Data Report, Paper No. 990389, Transportation Research Board Annual Meeting*.
- Puppala AJ, and Hanchanloet S (1999) Evaluation of a new Chemical (SA-44/LS-40) Treatment Method on Strength and Resilient Properties of a Cohesive Soil. *Technical Data Report, Paper No. 990389, Transportation Research Board Annual Meeting*.
- Rao SM and Thyagaraj T (2003) Lime slurry stabilization of an expansive soil. *Proceedings of the Institution of Civil Engineers – Geotechnical Engineering* 156(3): 139–146.
- Richards BG (1990) *Footings for small and domestic structures*. Linn Education and Training Services, Brisbane.
- Saride S, Puppala AJ and Chikyala SR (2013). Swell-shrink and strength behaviors of lime and cement stabilized expansive organic clays. *Applied Clay Science*, (85), 39-45.
- Sarkar G, Islam R, Alamgir M and Rokonzaman (2012) Study on the Geotechnical Properties of Cement based Composite Fine-grained Soil, *International Journal of Advanced Structures and Geotechnical Engineering*, 1(2): 42-49.
- Sarkanen KV and Ludwig CH (1971) *Lignins: Occurrence, Formation, Structure and Reactions*, Wiley, New York.
- Seed HB, Woodard R J and Lundgren R (1962) Prediction of swelling potential for compacted clays, *Journal of American Society of Civil Engineers, Soil Mechanics and Foundations Division*, 88(SM3): 53– 87.
- Skempton AW (1953) The colloidal activity of clays, *Proceedings 3rd International Conference of Soil Mechanics and Foundation Engineering*, Switzerland, (1) 57-61.
- Thakur VK, Thakur MK, Raghavan P, and Kessler, MR (2014) Progress in green polymer composites from lignin for multifunctional applications: a review< *ACS Sustain Chem Eng* 2: 1072-1092.

- Tingle JS, and Santoni RL (2003) Stabilization of clay soils with non-traditional additives. *Transportation Research Record No. 1819, National Research Council, Washington*, 72.
- Vinod JS, Indraratna B and Mahamud MAA (2010) Stabilisation of an erodible soil using a chemical admixture, *Proc., ICE – Ground Improvement*, 163(1), 43–51.
- Zhao XB, Su FY and Xing XH (2005) *Oxidation of sodium lignosulphonate by peracetic acid*. *J. Tsinghua University of Science and Technology*, 45(9): 1244-1247.

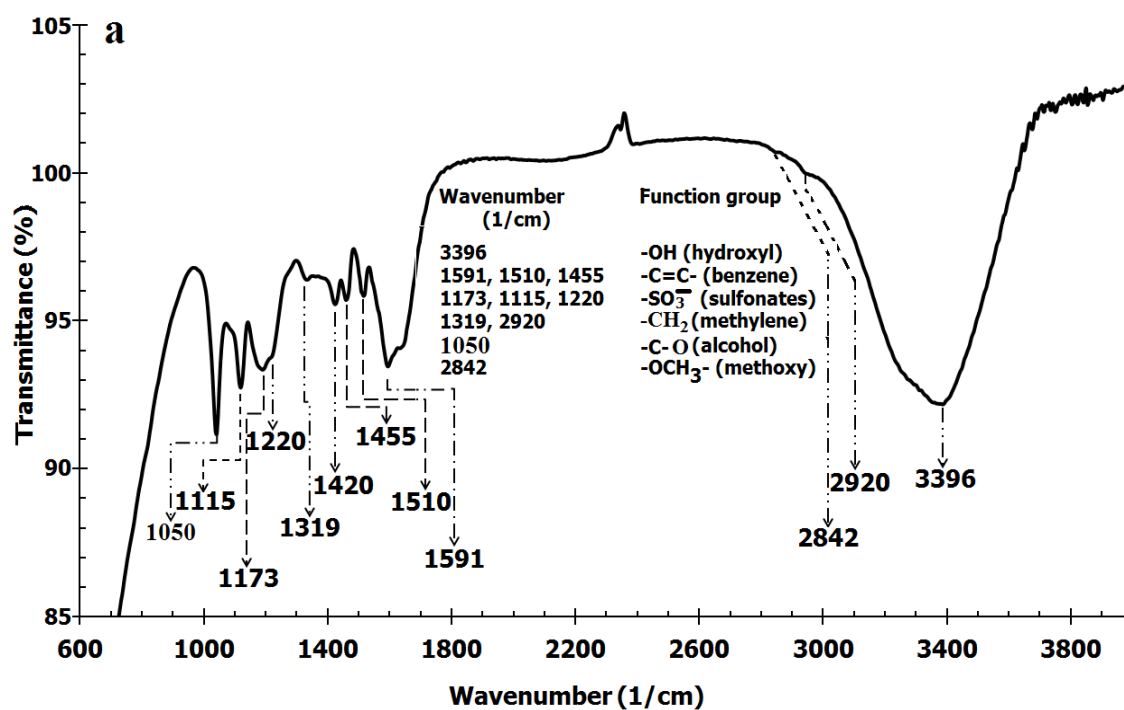
Table 1: Basic index properties and chemical composition of the remoulded expansive soil

Soil property	Value
Colour	Brown
Depth of sampling (m)	0.25 – 0.70
Natural moisture content (%)	8
Specific gravity	2.70
Liquid limit (%)	91
Plastic limit (%)	40
Plasticity index (%)	51
Shrinkage limit (%)	9
Classification (USCS) (unified soil classification system)	CH
Optimum moisture content (OMC, %)	37
Maximum dry unit weight (MDUW, kN/m ³)	13.1
Sand content (%)	9
Silt content (%)	53.6
Clay content (%)	36.0
Soil activity	1.42
Soil pH	7.43
% swell of remoulded sample under 7kPa @ OMC & MDUW	6
Corrected swelling pressure (kPa) of remoulded sample at OMC and MDUW	220
Unconfined compressive strength (kPa) of remoulded sample at OMC and MDUW	270
Compound	Mass (%)
Silica (SiO ₂)	55.1
Alumina (Al ₂ O ₃)	19.49
Ferric oxide (Fe ₂ O ₃)	13.33
Calcium (CaO)	2.69
Magnesium (MgO)	3.56
Titanium (TiO ₂)	0.16
Sodium (Na ₂ O)	0.21
Loss on ignition	5.45

NOTE: Swelling pressure of soil was determined using the constant volume method (ASTM D4546) and the construction procedure to correct the effect of sample disturbance was after; Fredlund, (1983).

Table 2: Elemental composition of LS stabilizer from EDS analysis

Element	Mass (%)
Carbon (C) mass in LS	47.14
Oxygen (O) mass in LS	23.59
Sulfur (S) mass in LS	12.98
Calcium (Ca) mass in LS	14.55
Aluminium (Al) mass in LS	1.02
Magnesium (Mg) mass in LS	0.21
Potassium (K) mass in LS	0.08
Sodium (Na) mass in LS	0.02



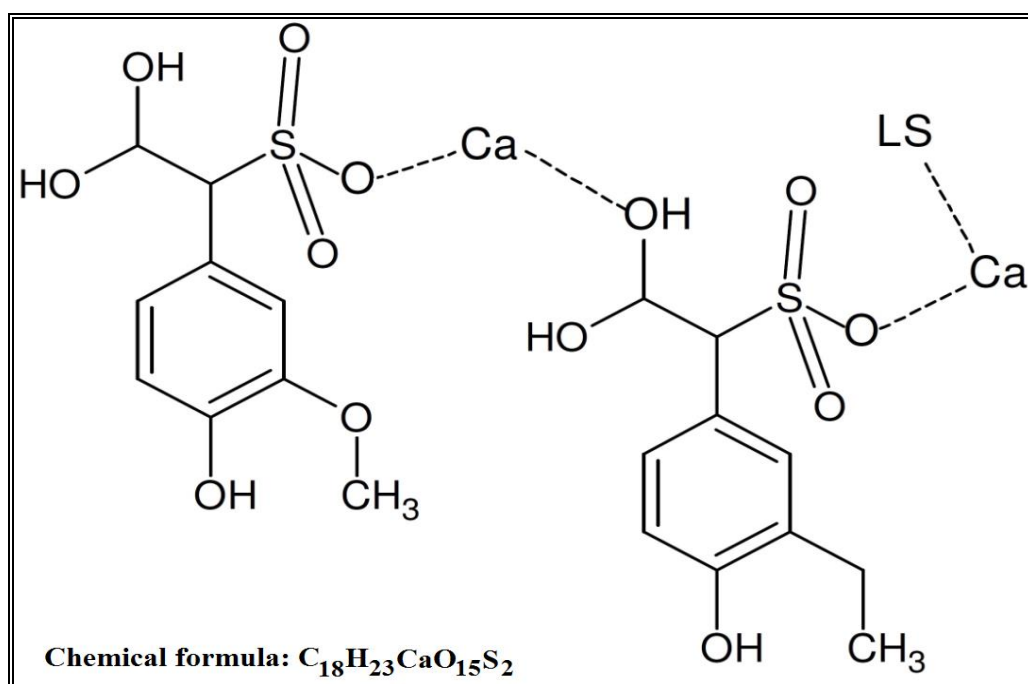


Fig 1. (a) FTIR pattern of LS admixture and (b) chemical structure of LS admixture

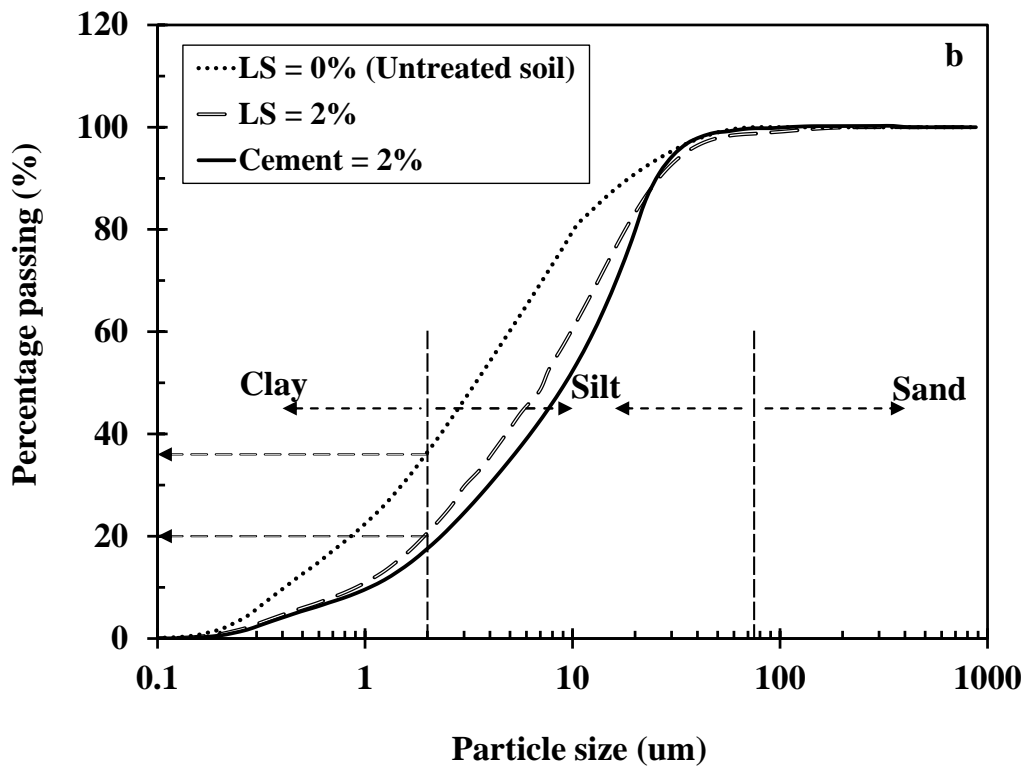
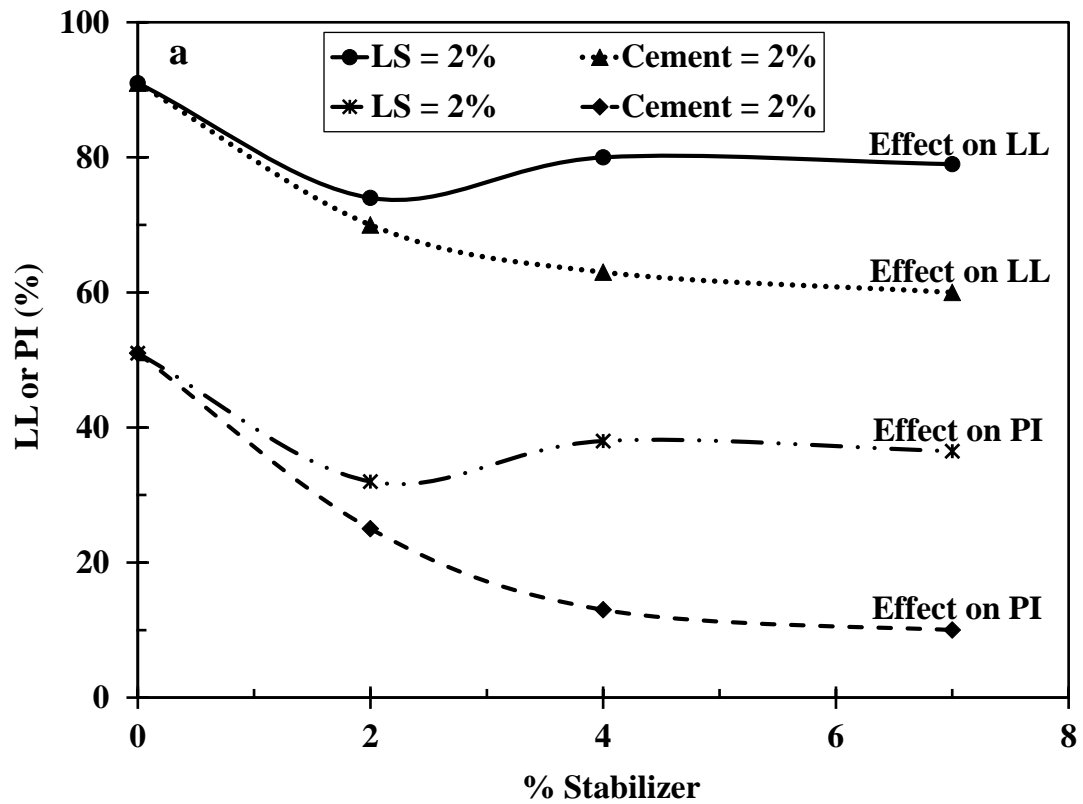


Fig 2. (a) Effect of LS content on the consistency limits of the remoulded expansive soil and (b) chemical treatment effect on the particle size distribution of the expansive soil

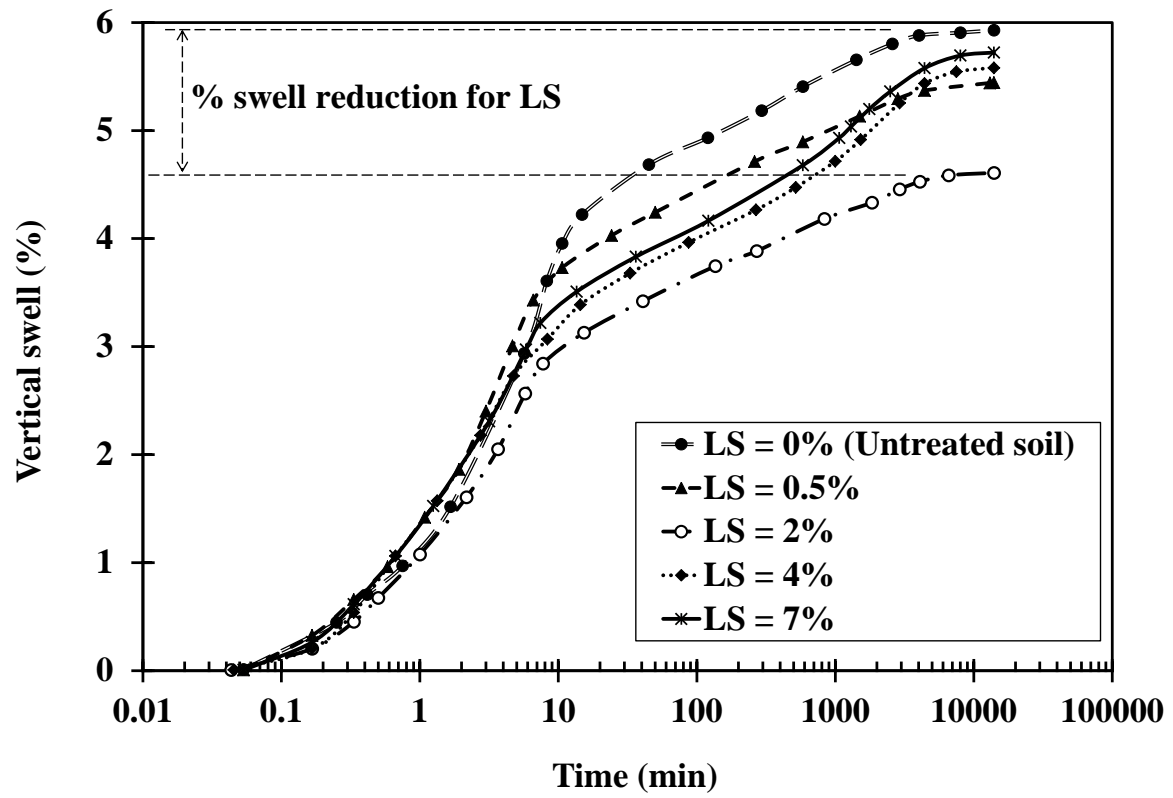


Fig 3. Effect of LS content on the percent swell with time

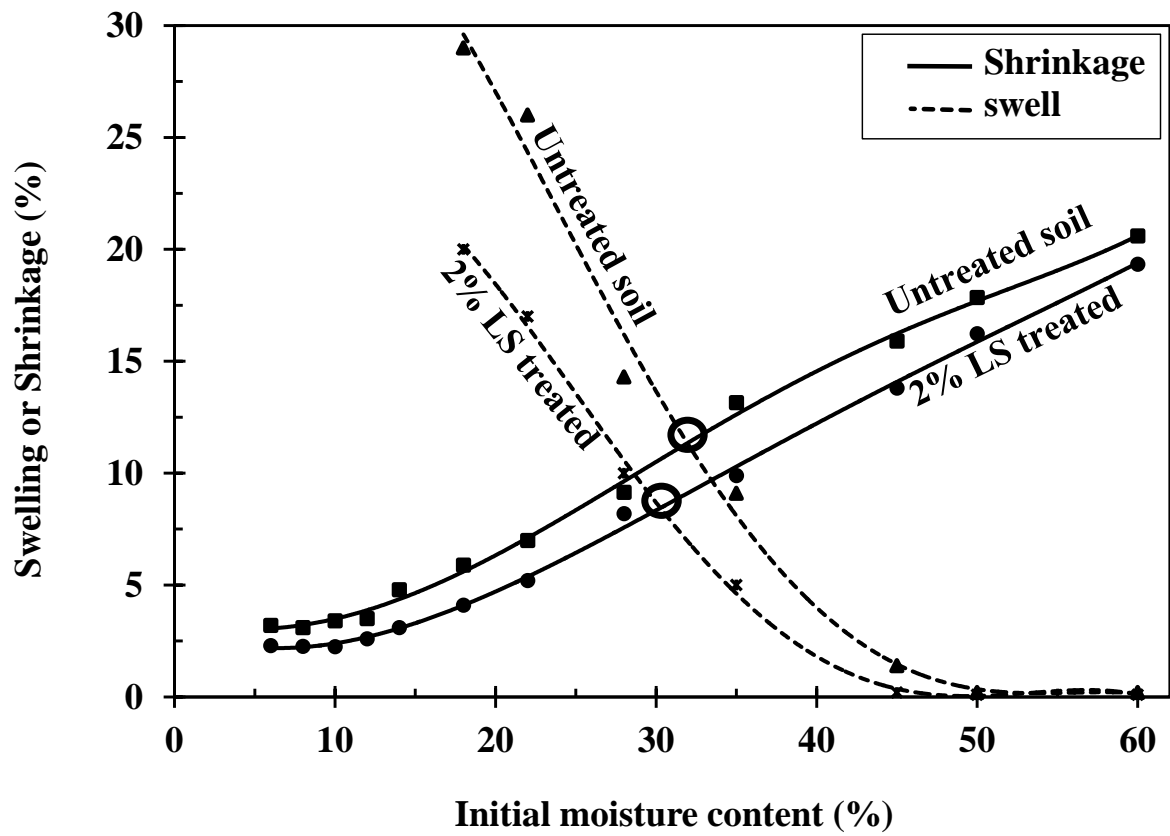


Fig 4: Effect of 2% LS treatment on the volumetric strain of the remoulded expansive soil

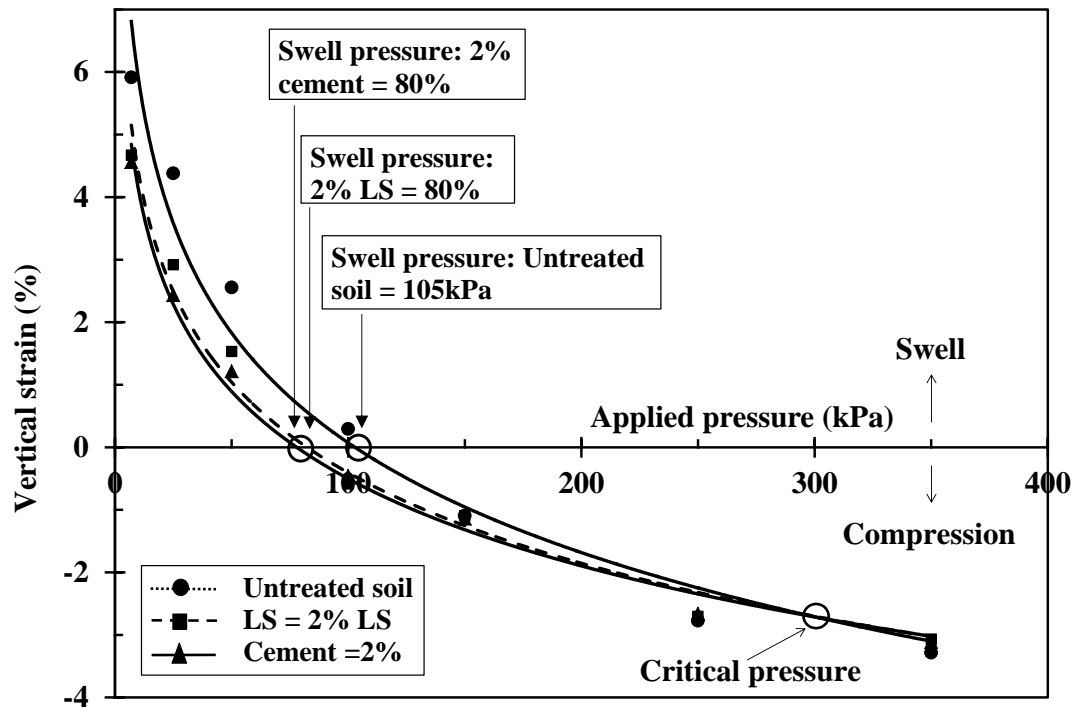


Fig 5. Constant volume swelling pressure measurement of untreated and treated expansive soil.

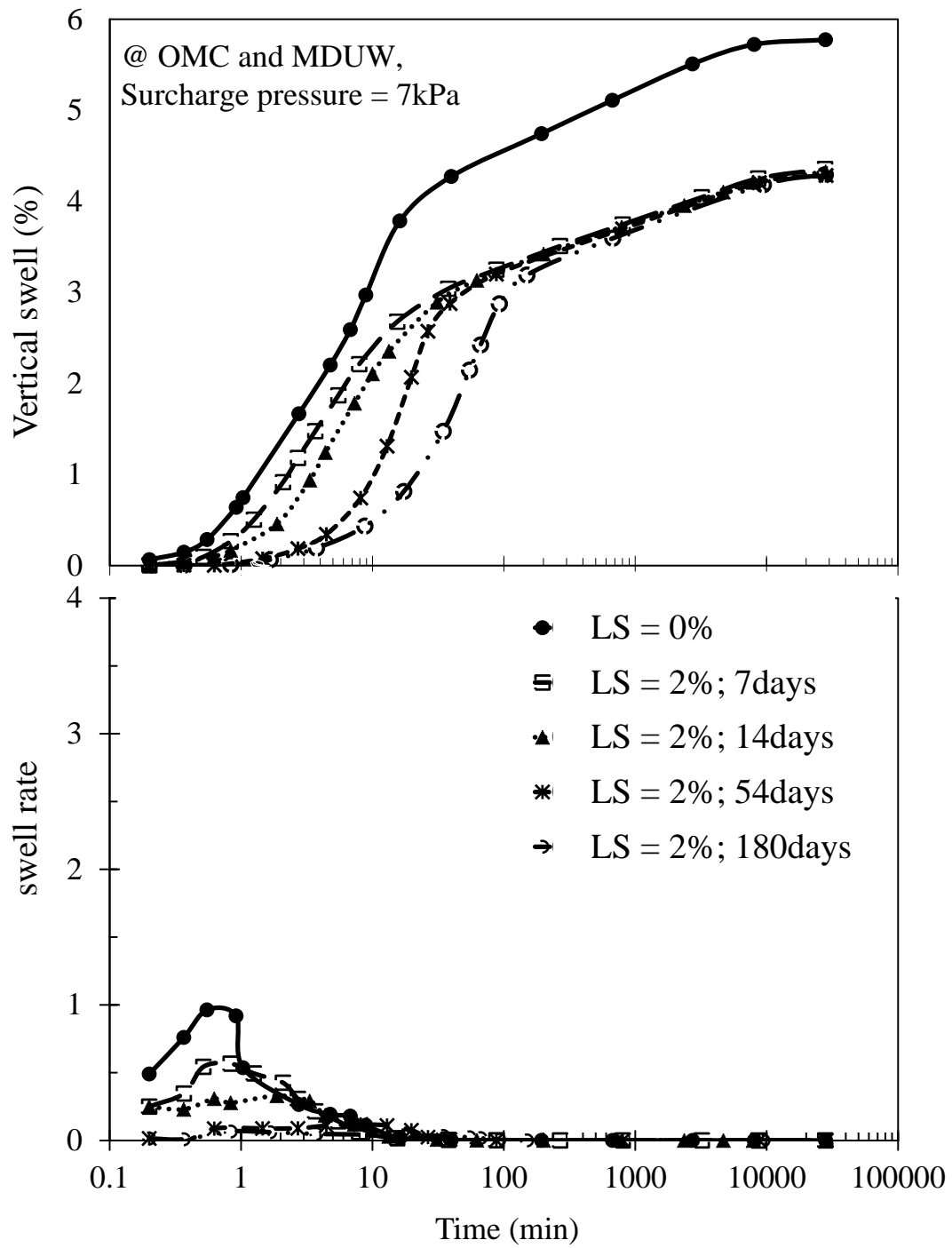


Fig 6. Time-dependent swell behaviour of L- treated expansive soil

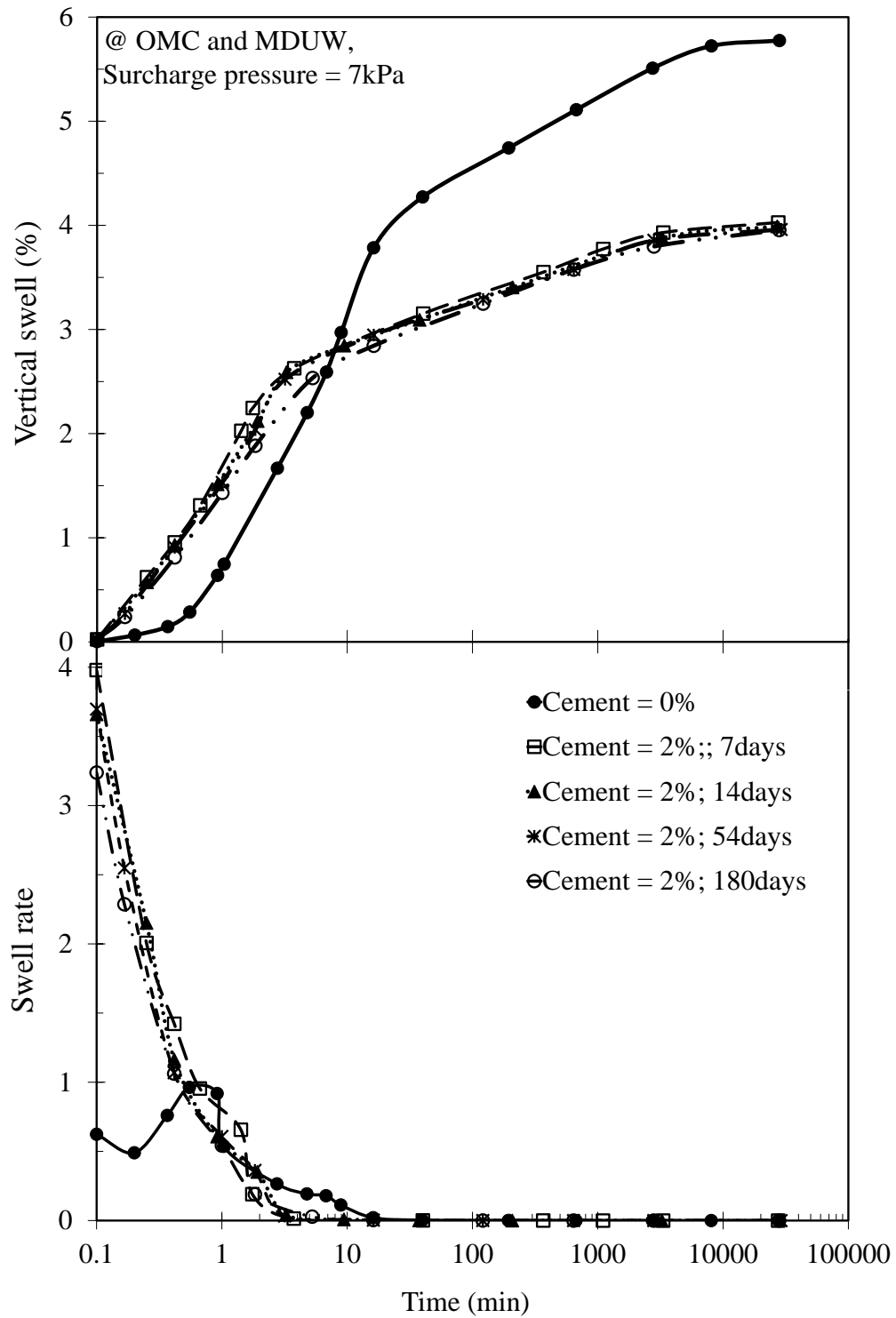


Fig 7. Time-dependent behaviour of cement-treated expansive soil

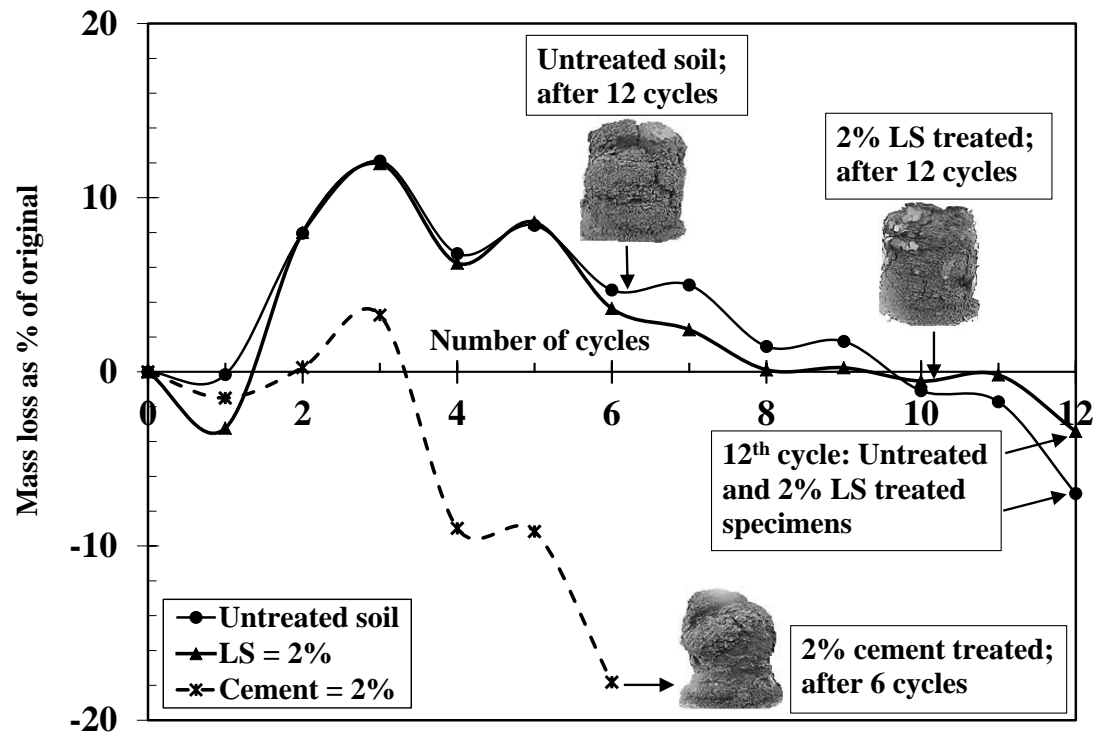


Fig 8. Effect of 2% LS content on mass loss in freeze-thaw durability test

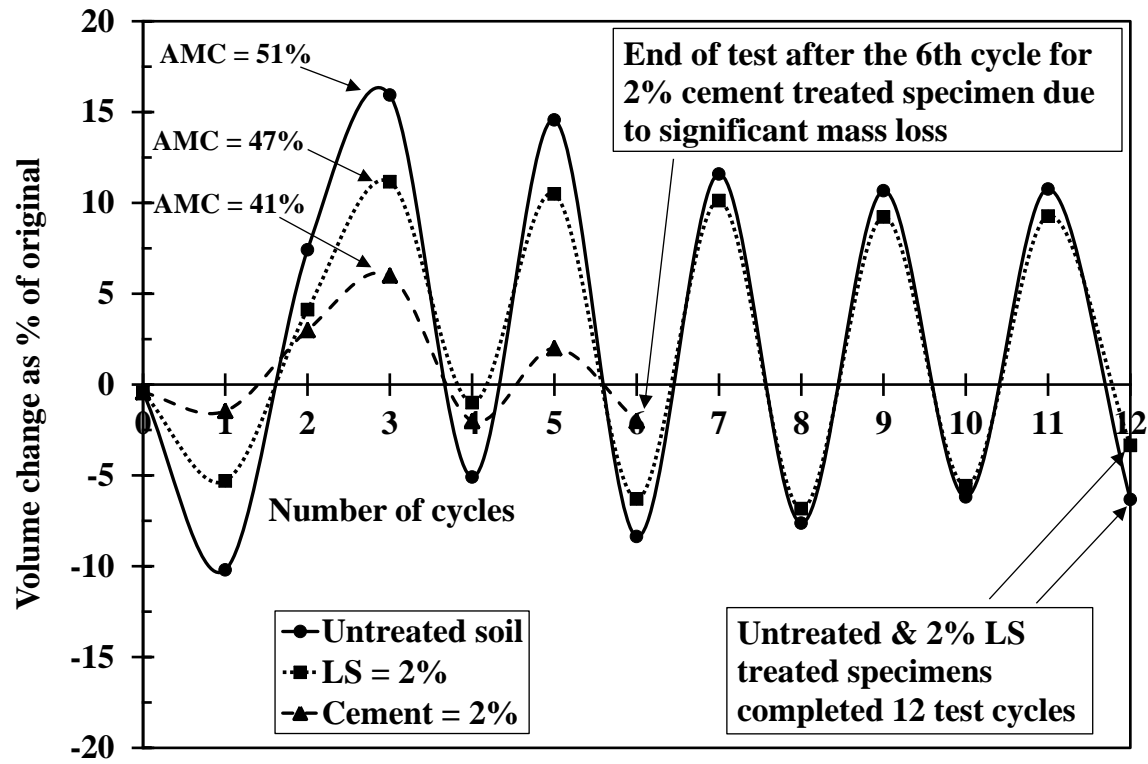


Fig 9. Effect of 2% LS content on moisture content at the end of each cycle in freeze-thaw durability test. AMC = adsorbed moisture content

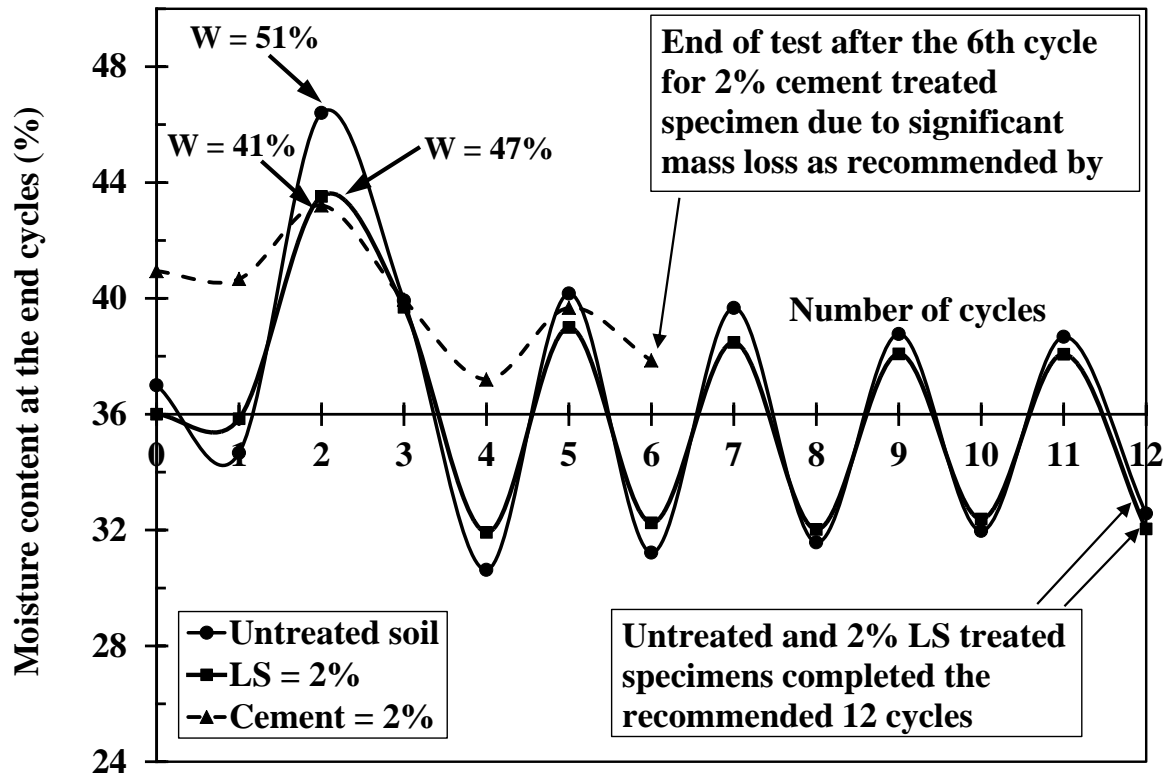


Fig 10. Effect of 2% LS content on volumetric strain in freeze-thaw durability test

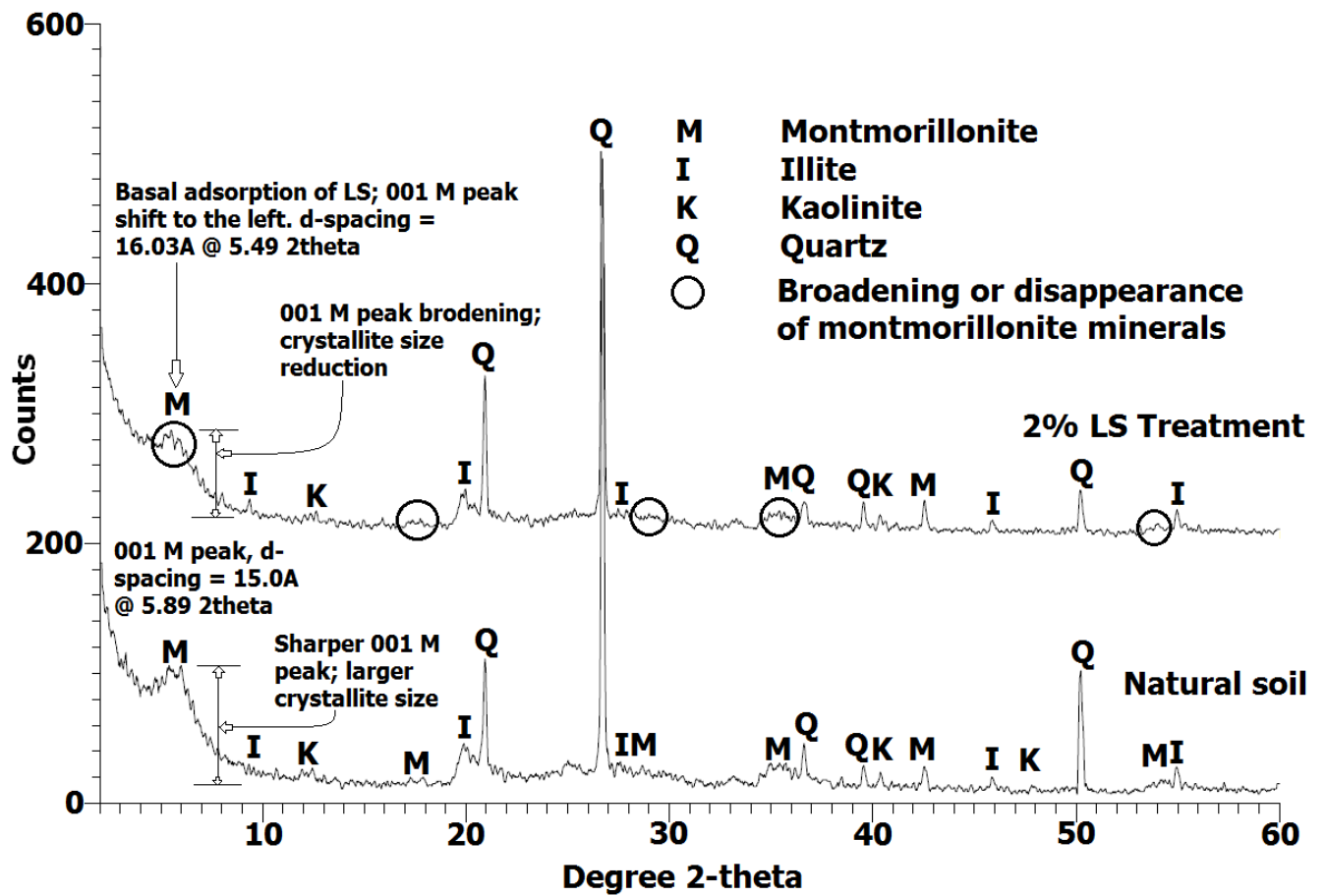


Fig 11. XRD diffractograms of untreated and 2% LS-treated expansive soil

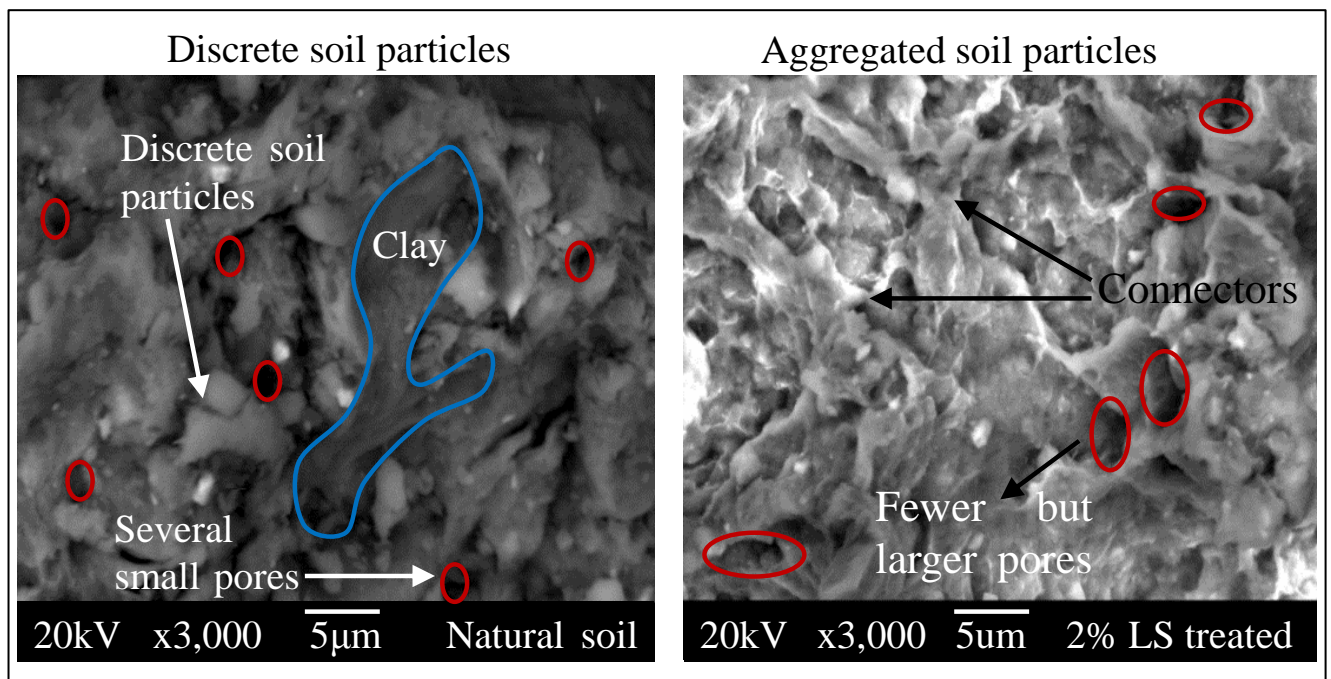


Fig 12. SEM micrographs of remoulded untreated and 2% LS-treated expansive soil